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Salt precipitation due to supercritical gas injection: I. Capillary-driven flow in unimodal sandstone

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ABSTRACT

Drying and salt precipitation in geological formations can have serious consequences for upstream operations in terms of injectivity and productivity. Here we investigate the consequences of supercritical CO_2 injection in sandstones. The reported findings are directly relevant for CO_2 sequestration and acid–gas injection operations, but might also be of interest to a broader community dealing with drying and capillary phenomena.

By injecting dry supercritical CO_2 into brine-saturated sandstone, we investigate the drying process and the associated precipitation of salts in a capillary-pressure-dominated flow regime. Precipitation patterns were recorded during the drying process by means of μCT scanning. The experimental results and numerical simulations show that under a critical flow rate salt precipitates with an inhomogeneous spatial distribution because of brine and solutes being transported in counter-current flow upstream where salt eventually precipitates. A substantial impairment of the absolute permeability has been found, but despite high local salt accumulation, the effective CO_2 permeability increased during all experiments. This phenomenon is a result of the observed microscopic precipitation pattern and eventually the resulting $K(\phi)$ relationship.

The findings in this paper are related to unimodal sandstone. In a companion paper (Ott et al., 2014) we present data on the distinctly different consequences of salt precipitation in dual- or multi-porosity rocks.

1. Introduction

Drying of porous media is an important topic in many industrial processes, in soil science and for upstream operations such as gas injection and production into and from geological formation. Drying of saline formations will cause precipitation of salt initially dissolved in the brine. This can negatively affect the performance of injection and production wells and can even lead to well clogging, which is a serious risk for such operations. In this paper we consider large-scale geological storage of CO_2, originating from anthropogenic sources like fossil-fueled power plants or contaminated gas production, in order to reduce CO_2 emissions. Deep saline aquifers and depleted oil and gas fields are potential subsurface deposits for that purpose (IPCC, 2005; Bachu and Gunter, 2004).

If dry or under-saturated, supercritical (SC) CO_2 is injected into water-bearing geological formations like saline aquifers, water is removed either by viscous displacement of the aqueous phase or by evaporation/dissolution of water in CO_2 and subsequent advection in the injected CO_2-rich phase. Both mechanisms act in parallel, but while advection of the aqueous phase decreases with increasing CO_2 saturation (diminished mobility), evaporation becomes increasingly important as the aqueous phase becomes immobile. Below residual water saturation, only evaporation takes place and the formation dries out if no additional source of water is available. If water evaporates, the salts originally present in the water are left behind. In highly saline formations, the amount of salt that potentially precipitates per unit volume can be quite substantial. The volumes depend on brine salinity, and the transport of solutes and water in the reservoir. Since fluid saturations and flow rates close to the well bore cover a large range as functions of space and time, there are no easy answers to the questions whether, where and how salt precipitates and how precipitation affects injectivity. The questions that need to be addressed are about the mechanisms of solute

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transport on a macroscopic scale that determine the macroscopic distribution of salt, and the salt distribution on a pore scale that determines how the permeability is affected as function of porosity reduction.

Even though salt precipitation in the vicinity of gas production wells are generally considered as issue, there are not many studies published referring to well data. The frequently cited paper is that of Kleinitz et al. (2001). Probably due to the limited number of industrial-scale CO_2 sequestration projects, there are no well data available referring to salt precipitation during CO_2 injection.

The lack of well studies renders a proper problem statement difficult and we rely on numerical simulations and analytical models that are available in literature. Here, we only aim to highlight a few key studies on precipitation due to CO_2 injection relating to the problem discussed in this paper. In a series of publications, Pruess et al. presented numerical simulations of CO_2 injection in saline aquifers, investigating the fundamental aspects of formation dry-out and salt precipitation (Pruess and García, 2002; Pruess and Müller, 2009). The simulations were performed with a single injection well in idealized 1D and 2D radial geometries. The authors observed that precipitation occurs only in a narrow dry-out zone confined to a few meters around the injection well. The solid salt saturation in this zone has been found to be constant independent of the injection rate. 2D radial simulations were carried out to explore gravity effects. In contrast to the 1D-radial scenario, gravity in combination with capillary-driven flow lead to more heterogeneous precipitation, with a maximum observed solid-salt saturation of more than 20%.

Giorgis et al. (2007) performed field-scale simulation in radial geometry. The authors found that the amount of precipitate depends on brine mobility and can be high if there is a capillary-gradient driven brine flow in the direction of the well bore. The authors have further shown that the injection rate is an important factor in controlling precipitation process and in avoiding or allowing complete clogging of the formation. In their simulations, solid salt saturations of locally more than 60% have been reached.

However, field-scale simulations require input on flow physics and thermodynamics such as the $K(\phi)$ relationship and the mass transfer rates between the fluid phases. As we will show in this paper and in Ott et al. (2014), these parameters are of microscopic origin and need to be determined by laboratory experiments. The quality of the input is critical for reliable simulations as several studies suggest that a modest change in porosity might lead to a serious reduction in permeability. The respective literature is diverse in mechanisms and there are not many studies dealing with fluid-transport induced porosity changes. The discussed mechanisms range from the porosity variation due to lithology/rock type (Pape et al., 1999; Ehrenberg and Nadeau, 2005) via mechanical compaction (Wyble, 1958; Schutjens et al., 2004) to silica dissolution and precipitation in geothermal systems in single-phase flow (Xu et al., 2004b) to drying processes, i.e. in two-phase flow as discussed in the present paper. Even if usually described by power laws, it cannot be expected that the $K(\phi)$ relationships resulting from different mechanisms are comparable – i.e. process-independent – and generally applicable.

There are only a few studies on flow-through drying available, which are relevant for CO_2 storage. Zuluaga et al. investigated vaporization and salt precipitation in sand packs and sandstone for gas production wells (Zuluaga and Monsalve, 2001; Zuluaga et al., 2001). At the GHGT-10 in Amsterdam 2010, two experimental studies on dry CO_2 injection have been presented. The experiments have been performed in sandstone in realistic storage conditions addressing capillary-driven solute transport, the condition of counter current flow (Ott et al., 2011b), and a permeability porosity relationship (Bacci et al., 2011). Recently, Peyssona et al. (2014) and Andre et al. (2014) investigated the drying process by nitrogen injection in sandstone. The data have been used to benchmark a numerical simulation tool for field-scale modeling of CO_2 injection. Ott et al. (2011b) pointed out that modeling of vaporization by an equilibrium approach is not sufficient to describe core flood experiments. Roels et al. (2014) performed core flood experiments and succeeded in the description of the saturation profiles by a kinetic approach.

In this paper we show results of core-flood experiments that were presented at the SCA conference in Halifax and at the GHGT10 in Amsterdam in 2010 (Ott et al., 2010, 2011b). The experiments were performed to investigate the drying process and the impact of salt precipitation on flow, i.e. the salt distribution and the $K(\phi)$ relationship. For this we injected dry SC CO_2 in brine-saturated siliciclastic sandstone. The experiments were carried out at flow rates realistic for near-well-bore flow and at realistic thermodynamic conditions. During injection, spatial and time evolution of saturation changes were monitored by means of micro computed tomography (μCT). The results in this paper are based on a number of experiments showing precipitation profiles with different degrees of heterogeneity on a macroscopic scale. For quantification we discuss two of these experiments showing the largest and the smallest spatial variation of salt saturation after dry-out. We refer to the cases as the heterogeneous and the homogeneous case, respectively. We explain the mechanisms that lead to the observed heterogeneous distribution of the precipitate by means of numerical simulations.

In addition to the saturation profiles, changes in absolute permeability and effective CO_2 permeability were monitored. From this data, we extract the permeability/porosity relationship ($K(\phi)$). We explain the observed mild permeability reduction by the microscopic distribution of the salt with respect to the observed CO_2 flow channels. Furthermore, we discuss the results in terms of precipitation in single-phase and two-phase flow situations.

2. Materials and methods

The experiments were carried out in a core flood setup designed for flooding with volatile and reactive fluids as sketched in Fig. 1. A detailed description of the unit can be found elsewhere (Ott et al., 2012). In the following, only a brief description of the elements will be given that are of relevance for the experiments here presented.

The flow experiments were performed in vertical geometry, with fluids being injected from top to bottom. The samples were embedded in polycarbonate and placed in a carbon-fiber based core holder – both are materials with low X-ray attenuation coefficients. The core holder is placed in a μCT scanner for in situ 3D imaging of the rock-fluid system. CT imaging allows to determine fluid saturations and changes of the rock matrix due to salt precipitation. The unit is equipped with two feed sections for liquids and liquefied gas injection. The CO_2 feed pump was held at 3 °C during the experiments – liquid CO_2 was injected and heated to experimental temperature and to the respective SC state in the injection lines. The density difference has been taken into account for the indicated flow rates. From flow rates and the differential pressure measurement (ΔP), the absolute (K) and effective permeability ($K \times k_{rel}$) were derived on line.

The experiments were performed on Berea sandstone with an average permeability of 500 mD and 22% porosity. The samples were drilled from the same block and were small in cross section and volume (1 cm ø and 5 cm length) to obtain representative flow rates and to reduce the experimental time to complete dry-out. The mineralogy of Berea is dominated by quartz with some K-feldspar, kaolinite, and minor amounts of other clay minerals as determined by eSEM/EDX. The rock samples were pre-saturated with NaCl-based high-salinity brine: 20 wt% NaCl and 2 wt% CsCl.